

## SCIENCE FOR CERAMIC PRODUCTION

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### TIME AND STRAIN TO RUPTURE OF DENSE POLYCRYSTALLINE OXIDE CERAMIC AT TEMPERATURES TO 1600°C

V. S. Bakunov,<sup>1</sup> E. S. Lukin,<sup>2</sup> and É. P. Sysoev<sup>3</sup>

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The indices of long-time strength (time and strain to rupture) of densely sintered oxide ceramic in the temperature interval 1400 – 1600°C under loads to 60 MPa are determined. The characteristics of creep and creep rupture are interrelated: irrespective of the size of the crystals in the ceramic and the testing conditions the product of the steady creep rate and the time to rupture is a constant, i.e., the strain to rupture is a constant. Therefore, the service life of materials can be determined from measurements of the creep rate.

**Key words:** densely sintered ceramic, periclase, corundum, aluminum-magnesium spinel, service life, creep, time to rupture, strain.

The particulars of strain and rupture processes in densely sintered oxide ceramic at temperatures to 1600°C, i.e., somewhat above the temperature of the brittle-to-plastic transition, are examined in [1 – 3]. It is shown in [1] that in the entire temperature interval of the plastic behavior of ceramics the main mechanism of deformation is diffusive-viscous flow, which can be accompanied by diffusive movement of dislocations (climb) and diffusive displacement of crystal boundaries.

As established in [4], a period of nonstationary strain can be observed in diffusive-viscous flow of polycrystalline bodies. It results from the local strain from stress relaxation on micro defects being added to the flow strain. The micro defects formed under the nonequilibrium conditions of cooling of materials from the high temperatures at which they are fired.

When the samples are heated to the testing temperature these defects annihilate all the more completely the higher the temperature reached. In the course of testing they continue to annihilate until stationary creep is reached. Nonequilibrium defects remain in a ceramic in the entire temperature interval of its brittle behavior. Then, when a certain degree of

plasticity is reached, they are removed via annihilation, a process that is thermally activated. Thus, the more the testing temperature exceeds the temperature of the brittle-to-plastic transition, the lower the content of nonequilibrium defects. The contribution of the nonstationary strain to the overall rupture strain (strain-to-rupture) decreases.

The rupture of a ceramic, specifically, in bending, also occurs by the diffusion pathway during steady creep (i.e., creep at a constant rate) as a result of, first, vacancy formation in sections of the stretched zone that are tied to the crystal boundaries and then coagulation of vacancies on the boundaries [1]. It was found that the strain and rupture processes are not the same in different materials and depend on the crystal-chemical particulars of their structure [2]. It was shown for periclase ceramic that the deformation and rupture processes are interrelated [3]: the product of the time-to-rupture and the steady creep rate is a constant and independent of the testing conditions (temperature and load) as well as the crystal size.

The present work continues [3] in the context of expanding the range of materials.

The purpose of the present article is to present the results of experiments undertaken to determine the indices of long-time strength of other densely sintered materials (corundum, aluminum-magnesium spinel) as well as to determine the particulars of the behavior of polycrystalline ceramic under a mechanical load at temperatures to 1600°C.

<sup>1</sup> Joint Institute for High-Temperature Research, Russian Academy of Sciences, Moscow, Russia (e-mail: bacsenok@mail.ru).

<sup>2</sup> D. I. Mendeleev Russian University of Chemical Technology, Moscow, Russia.

<sup>3</sup> Vladimir State University, Vladimir, Russia.

**TABLE 1.** Characteristics of the Experimental Samples

Body index	Type of ceramic	Predominate crystal size, $\mu\text{m}$	Apparent density, $\text{g/cm}^3$	True porosity, %	Strength in bending, MPa, at temperature, °C				
					20	1400	1450	1500	1550
K-15	$\text{Al}_2\text{O}_3$	15	3.90	2.5	150	70	56	52	50
K-35	$\text{Al}_2\text{O}_3$	35	3.90	2.5	125	60	54	51	42
ShP-10	$\text{MgAl}_2\text{O}_4$	10	3.49	2.5	150	35	22	18	15
ShP-25	$\text{MgAl}_2\text{O}_4$	25	3.50	2.2	120	30	20	15	13
P-12	$\text{MgO}$	12	3.47	3.0	120	65	52	45	40
P-25	$\text{MgO}$	25	3.50	2.2	105	60	73	62	55

The long-time strength was determined at constant stresses in the range 5 – 30 MPa for spinel and 20 – 50 MPa for corundum. The particulars of the methods used are presented in [1, 2] and some characteristics are presented in Table 1. We note that judging from the data in the table the interval of measurement temperatures (1400 – 1550°C) lies somewhat above the brittle-to-plastic transition temperature, since here the strength of the materials decreases monotonically depending on the heating.

The service life characteristics (time  $\tau_r$  and strain  $\varepsilon_r$  to rupture) were determined from the experiments. Initially, their dependences on the testing conditions (load and temperature) for two types of samples differing by the size of the crystals were examined. Next, the relation between the characteristics of the rupture process in the materials and the steady creep rate  $\dot{\varepsilon}_s$  was determined.

Judging from [1, 2] as well as the experimental results, the behavior of each type of ceramic is highly individual. This is probably associated with the particulars of their crystal lattice structures. For this reason the data are presented separately for each type, after which the general laws of their behavior under a load are formulated. The experimental results for periclase ceramic were also taken into account in the analysis [3].

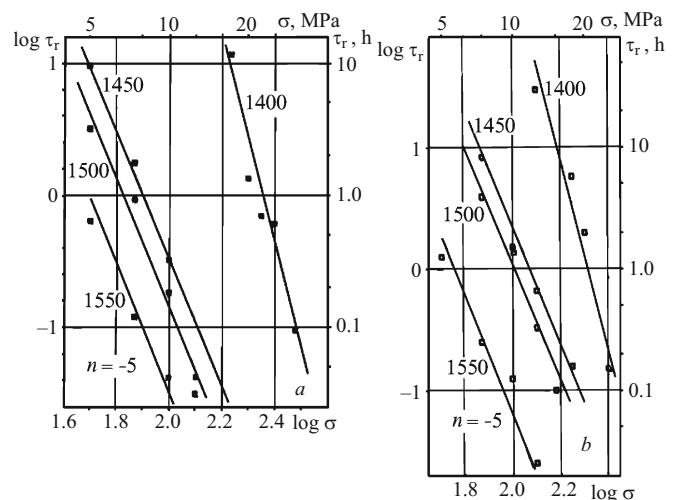
*Spinel-based ceramic.* Both experimental samples (ShP-12 and ShP-25) show at temperature 1400°C the longest time-to-rupture under identical stresses. A large variance was observed in the data, which is probably due to the very low measured flow deformation. The temperature 1400°C probably corresponds to the onset of the brittle-to-plastic transition, and here the effect of micro defects is large, especially for surface defects, which are effective stress concentrators. As the fluidity increases with subsequent heating the role of the concentrators is not so significant. For this reason mainly the results obtained at temperatures above 1400°C were taken into account in this work, since they are less subject to the random effect of uncontrollable defects formed in the samples cooling from the firing temperature in a flame furnace.

The time-to-rupture  $\tau_r$  versus the applied stress  $\sigma$  is presented in Fig. 1. One can see that in the experimental temperature range the time-to-rupture decreases with increasing

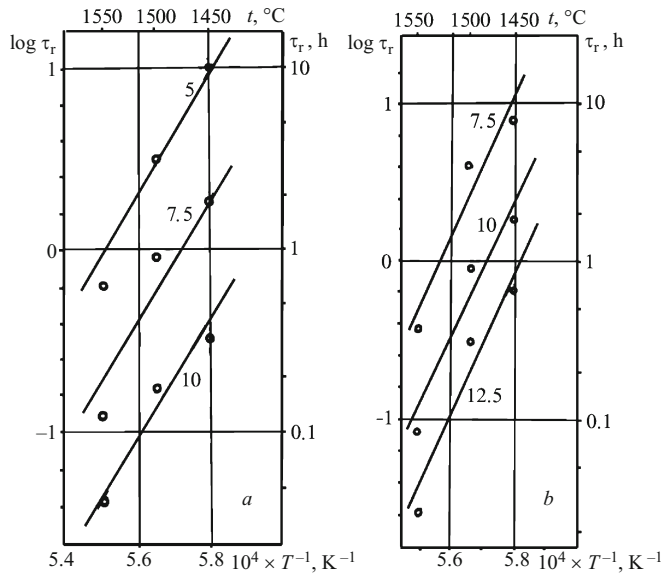
stress. In logarithmic coordinates the experimental points fall on straight lines with the same slope angle, whose tangent  $n \approx -5$  for the two types of samples studied. Therefore, at temperatures above 1400°C the mechanism of the processes remains constant as a function of temperature. Analytically, the function can be expressed in the form  $\tau_r = A\sigma^n$ , where  $n = -5$ ,  $A = \text{const}$  and  $A$  is determined mainly by the testing temperature. We note that the results obtained at temperature 1400°C, which also all fall on the same straight but with a larger slope ( $n = -8$ ), stand apart from the general dependence.

The time-to-rupture is a decreasing function of temperature  $T$  (Fig. 2):  $\tau_r = B \exp [-Q/(RT)]$ , where  $R$  is the gas constant and  $B = \text{const}$ . The activation energy  $Q$  calculated from the slope of the straight lines is the same for both types of ceramics and equals approximately 650 kJ/mol (close to the activation energy of creep 600 kJ/mol [2]). The results obtained at 1400°C stand apart from the general dependence.

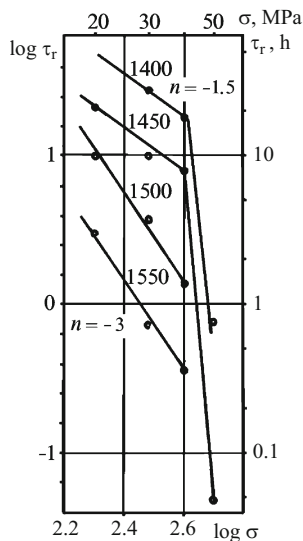
*Corundum ceramic.* As in the case of spinel ceramic, the longest time-to-rupture for both types of samples (K-15 and K-35) under identical stresses is observed at temperature 1400°C. Similarly, under these testing conditions a large



**Fig. 1.** Time-to-rupture  $\tau_r$  versus the stress  $\sigma$  for spinel ceramic samples: a) ShP-10; b) ShP-25. The temperature is indicated in °C.

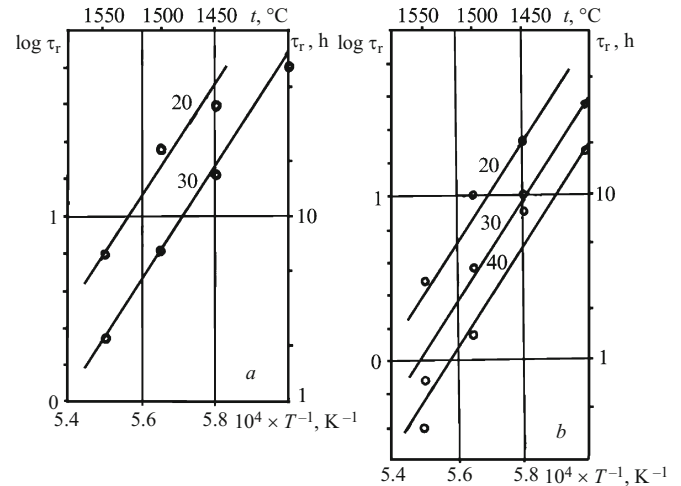


**Fig. 2.** Time-to-rupture  $\tau_r$  versus the temperature  $T$  for spinel ceramic samples: a) ShP-10; b) ShP-25. The load is indicated in MPa.



**Fig. 3.** Time-to-rupture  $\tau_r$  versus the stress  $\sigma$  for corundum ceramic samples K-15. The temperature is indicated in °C.

spread in the data was observed for very small measured values of the flow deformation. Most likely this behavior is associated with the states of samples only at the initial stage of the brittle-to-plastic transition. Apparently, a significant part of the measured deformation is associated with relaxation processes on micro defects, especially surface defects. It should be noted that large-crystal K-35 samples also exhibit similar behavior at 1450°C. Another feature of corundum ceramic is the presence of inflections in the time-to-rupture versus the stress (in logarithmic coordinates) and versus the temperature (in semilogarithmic coordinates); these dependences can be represented only in the form of piece-wise linear approximations. As an example, the results for the time-to-rupture of K-15 samples versus the applied load are



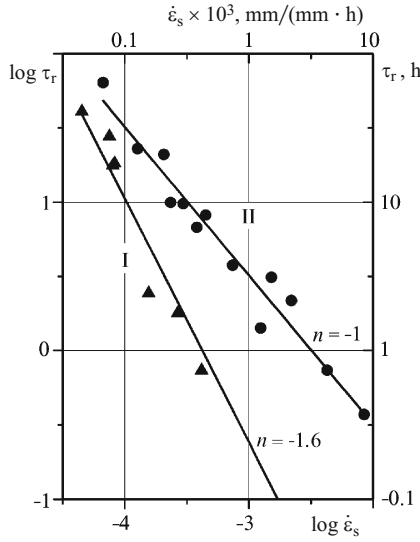
**Fig. 4.** Time-to-rupture  $\tau_r$  versus the temperature  $T$  for corundum ceramic samples: a) K-15; b) K-35. The load is indicated in MPa.

presented in Fig. 3. It is evident from Fig. 3 that the slope of the dependences changes as a function of the temperature and load. For example, for stresses below 40 MPa and temperatures 1400 – 1450°C the tangent of the slope angle of the straight is  $n \approx -1.5$ , but as temperature increases to 1500 – 1550°C  $n$  becomes  $n \approx -3$ . On the other hand, for loads above 40 MPa at temperatures 1400 – 1450°C the index  $n$  becomes  $-1.5$  or more.

The change in the behavior of samples under stress very close to the ultimate strength and temperatures somewhat above the region of the brittle-to-plastic transition is completely explainable, because here the mechanisms of brittle rupture, which differ from those of plastic (viscous) rupture, can operate here. On the basis of the data obtained and taking account of the results presented in [1 – 3] it can be assumed that the brittle-to-plastic transition occurs closer to the temperature 1450°C for corundum ceramic and 1400°C for spinel and somewhat below 1400°C for periclase.

On this basis, to determine the temperature dependence of the time-to-rupture for corundum ceramic we took account of the results corresponding to plastic rupture, i.e., relaxation on micro defects (especially surface defects) had no appreciable effect on the measured deformation (i.e., the deformation due to nonstationary creep is small).

The data obtained are presented in Fig. 4. For samples of each type (K-15, K-35) the experimental points in the coordinates chosen fall satisfactorily on a family of parallel straight lines for a given stress. Thus, the time-to-rupture  $\tau_r$  is a decreasing exponential function of temperature:  $\tau_r = B \exp [-Q/(RT)]$ , where  $R$  is the gas constant and  $B = \text{const}$ . The activation energy  $Q$  calculated from the slope of the straight lines is the same for both types of ceramics and equals approximately 550 kJ/mol (close to the activation energy of creep 600 kJ/mol [2]). We note that the results obtained at temperatures 1400 – 1450°C and stresses close to



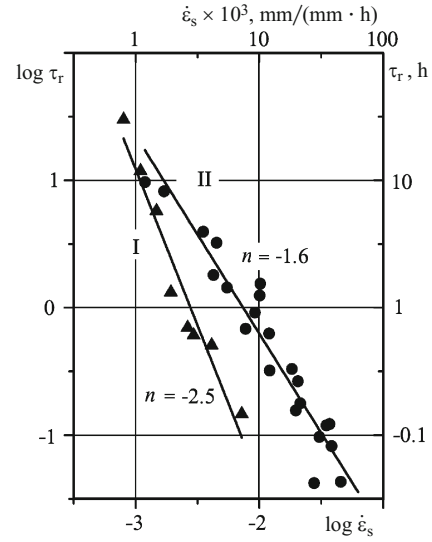
**Fig. 5.** Time-to-rupture  $\tau_r$  versus the rate of steady creep  $\dot{\epsilon}_s$  for corundum samples: I) at temperatures 1400 – 1450°C; II) main dependence.

the ultimate strength stand apart from this general dependence.

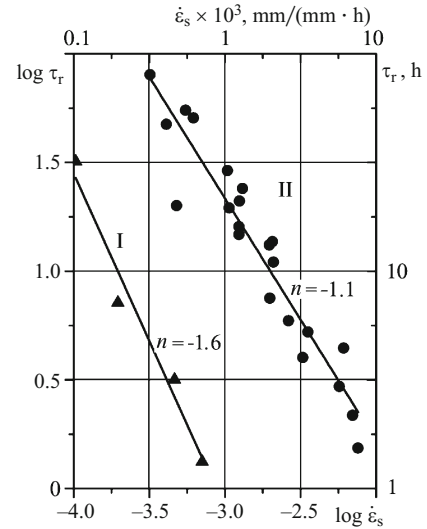
*Relation between the rupture characteristics and steady creep rate.* The time and strain-to-rupture versus the steady creep rate were constructed for the experimental ceramics on the basis of the data. It was assumed that, just as found in experiments on creep, certain analogies existed in the behavior of polycrystalline ceramic and metallic materials [5, 6]. The results of the present work were analyzed taking account of the data in [2, 3].

The experimental data on the time-to-rupture versus the steady creep rate for the experimental ceramics are presented in Figs. 5 – 7. For convenience the results for periclase ceramic are also presented [3]. The main feature of the plots draws our attention: for each of the experimental ceramics the results in the coordinates chosen fall on two straight lines differing in the slope angle. The bulk of the data represent an essentially linear function of  $\log \tau_r$  versus  $\log \dot{\epsilon}_s$  irrespective of the testing conditions and the size of the sample crystals: the tangent of the slope angle  $n$  of the straight lines is close in magnitude to  $n = -1$ . The linear relation between the logarithms of the quantities signifies a linear relation between the quantities themselves, which leads to the following very important conclusion from the data obtained: taking account of the negative exponent ( $-1$ ) it can be assumed that the product of the time-to-rupture and the steady creep rate of polycrystalline oxide ceramic is a constant irrespective of the testing conditions (load and temperature) and the size of the sample crystals.

The results for spinel and periclase at 1400°C and corundum at 1400 and 1450°C pertain to special groups: they fall on different straight lines. As found above, the deviation of the data from the main dependence is associated with the



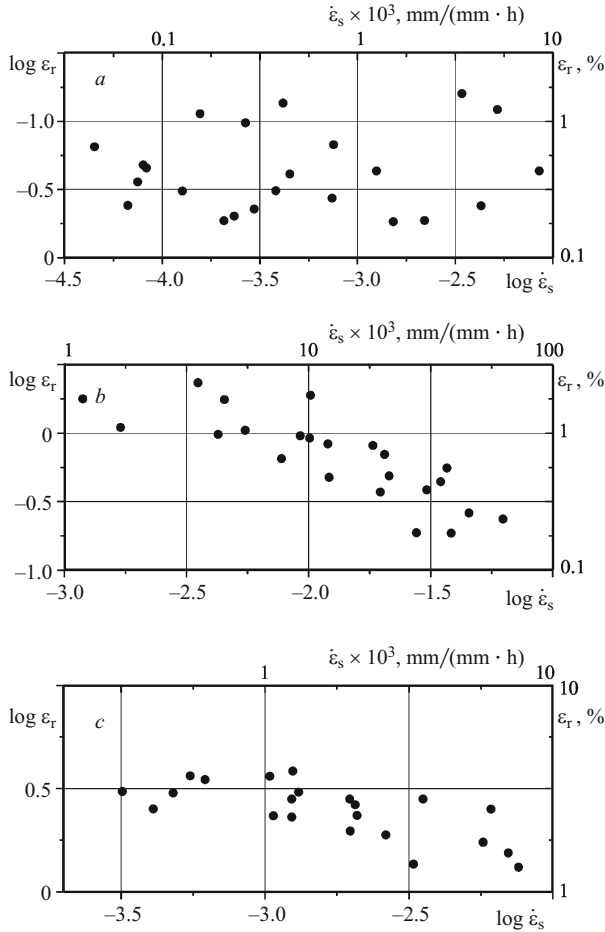
**Fig. 6.** Time-to-rupture  $\tau_r$  versus the rate of steady creep  $\dot{\epsilon}_s$  for spinel samples: I) at temperature 1400°C; II) main dependence.



**Fig. 7.** Time-to-rupture  $\tau_r$  versus the steady creep rate  $\dot{\epsilon}_s$  for periclase samples: I) at temperatures 1400°C; II) main dependence.

presence in the ceramic of nonequilibrium defects quenched-in when the ceramic cooled from high firing temperatures. Annealing of these defects (and the associated deformation) in the case of ceramic occurs only at temperatures above the region of the brittle-to-plastic transition. On the basis of the data in Figs. 5 – 7 and taking account of the results of [1] it can be assumed that the onset of this transition is observed at temperatures 1400 – 1450°C for corundum, 1400°C for spinel and somewhat higher than 1400°C for periclase.

In accordance with existing ideas [6] the strain-to-rupture  $\epsilon_r$  at constant load (i.e., during creep) in the general case can be represented as  $\epsilon_r = \epsilon_{ns} + \epsilon_s + \epsilon_{acc}$ , where  $\epsilon_{ns}$  is the



**Fig. 8.** Strain-to-rupture  $\varepsilon_r$  versus the steady creep rate  $\dot{\varepsilon}_s$  for ceramic: a) corundum; b) spinel; a) periclase.

strain during the nonstationary period,  $\varepsilon_s$  during the stationary period and  $\varepsilon_{acc}$  during the accelerated period. We have determined experimentally [1, 2] that, as a rule,  $\varepsilon_{acc} = 0$ ; very rare exceptions were found at high temperatures and under loads close to the ultimate strength of the material. For this reason, the relation  $\varepsilon_r = \varepsilon_{ns} + \varepsilon_s$  is usually used for polycrystalline ceramic. The quantity  $\varepsilon_s$  builds up by the mechanism of diffusive-viscous flow, i.e.,  $\varepsilon_s = \dot{\varepsilon}_s \tau_s$ , where  $\dot{\varepsilon}_s$  is the rate and  $\tau_s$  the time of the process; a large part of the total deformation occurs by this mechanism.

The deformation during the nonstationary period is usually small, and its role decreases with increasing temperature [1]. For constant testing conditions this deformation increases as a power-law function of time with exponent  $\alpha$ :  $\varepsilon_{ns} = \dot{\varepsilon}_{ns} \tau_{ns}^\alpha$ . Since the rate of nonstationary creep is higher than that of stationary creep, some average kinetic relation can be represented as a power-law function of the strain-to-rupture in time:  $\varepsilon_r = \dot{\varepsilon}_{av} \tau_c^m \tau_c^m$ , where  $m > 1$ . We note that the difference between  $m$  and 1 is determined by the contribution of the deformation during the nonstationary period to the total strain-to-rupture: the larger the difference, the

higher this contribution is. Hence it follows that for  $m \approx 1$  the strain-to-rupture is proportional to the time-to-rupture, and the coefficient of proportionality is the steady creep rate, i.e., in this case  $\varepsilon_r = \dot{\varepsilon}_s \tau_r$ .

Judging from the data obtained (see Figs. 5–7), in the experimental interval of temperatures the deformation of corundum and periclase, whose lattices are quite simple (trigonal and cubic, respectively), proceeds by the mechanism of viscous flow (exponent  $m \approx -1$ ). At the same time, the nonstationary creep associated with the annealing of nonequilibrium defects in a more complicated face-centered lattice makes an appreciable contribution to the deformation of spinel. For this reason the modulus of the exponent  $m$  is greater here and  $m = -1.6$ .

The dependence of the strain-to-rupture  $\varepsilon_r$  of the experimental materials on the steady creep rate  $\dot{\varepsilon}_s$  in logarithmic coordinates is presented in Fig. 8. As one can see from the plots, the strain-to-rupture is practically independent of the steady creep rate, but the variance in the data is large. At the same time the relations (Fig. 8) show a tendency for the strain to decrease with increasing creep rate. This could be associated on the one hand with the individual behavior of each sample near the brittle-to-plastic transition and on the other hand with a too small size of the experimental samples. In any case, additional research is needed to determine the mechanisms of the behavior of polycrystalline ceramic on heating under a load.

## CONCLUSIONS

The present work marks the conclusion (together with the publications [1–3]) in the general series of studies of the behavior of polycrystalline ceramic under a constant mechanical load (i.e., under creep conditions) in the range of temperatures somewhat above the temperature of the brittle-to-plastic transition with the applied stresses close to the ultimate strength of the ceramic at the given temperature of the tests.

It was determined that in the overwhelming majority of cases the samples rupture at the steady creep stage; accelerated creep was observed in singular cases and at stresses very close to the ultimate strength. Nonstationary creep, apparently associated with the relaxation on nonequilibrium defects, was observed at low temperatures near the elastic-plastic transition. In all probability, the brittle-to-plastic transition in polycrystalline ceramic is smeared as a function of the temperature over at least 100–150°C: according to the applied stress its position depends on the temperature and size of the sample crystals.

The creep and rupture of polycrystalline ceramic at temperatures above the range corresponding to the brittle-to-plastic transition occur by means of diffusive-viscous flow. For all experimental materials in this range the product of the time-to-rupture and the steady creep rate is a constant irrespective of the testing conditions (temperature and load) and

the structure of the materials, so that  $\dot{\epsilon}_s \tau_r = \text{const}$ . This shows that the deformation prior to rupture is also a constant.

For applications the main result is that the rupture characteristics ( $\tau_r$ ,  $\epsilon_r$ ) of a ceramic far from the region of its brittle-to-plastic behavior can be evaluated approximately according to the results of comparatively short-time experiments. In addition, there is no need for very prolonged and expensive tests for long-time strength.

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